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THERMAL EXPANSION OF BeO-SiC COMPOSITES

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For Space Systems Division and Ballistic Systems Division Air Force Systems Command

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ABSTRACT

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An investigation was made of the coefficient of thermal expansion to 1000°C for composite BeO-SiC bodies. Values were found to be in general agreement with those calculated from the equation proposed by Turner relating resultant expansion to the single component coefficients, weight fraction of the phases, phase density, and bulk modulus.

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I. INTRODUCTION

Re-entry systems, start-stop rocket nozzles, and other applications for refractories could be benefitted by improvement of the thermal shock resistance of the refractory material component. The amount of technical effort directed toward this end has been significant, especially since 1946 when an immediate need arose for jet turbine blade materials. Obviously, there is no single or simple system or panacea for designing shock resistance into various materials for diverse applications. A recommended approach is the delineation and characterization of the principal properties affecting thermal shock resistance, followed by the application of this science to the modifying, adjusting, and controlling of material properties.

These properties that inherently characterize thermal shock resistance for ceramics as reviewed by Hasselman (Ref. 1) are tensile strength, Young's Modulus, Poisson's ratio, coefficient of thermal expansion, thermal conductivity, thermal diffusivity, and emissivity. For polyphase systems, additional factors are the volume fraction, the size, shape, and distribution of particles, the porosity as it affects mechanical and thermal properties, the volume under stress, the effective surface energy required for crack propagation, and the elastic energy stored at fracture available for crack propagation.

In the consideration of the enhancement of thermal shock resistance of BeO bodies, the properties that can be most dramatically affected appear to be strength, thermal expansion, and emissivity. After reaching a near ultimate in property development for this pure material, an obvious recourse is to additional phases. Two methods considered for increasing strength are prestressing and reinforcement, both of which can be applied independent of micro-composition.

Addition of a secondary phase to a given matrix (BeO) appears essential for the enhancement of thermal expansion and/or emissivity and is, therefore, of special interest. By combining or compositing materials with

differing thermal expansion coefficients in a ceramic forming process, substantial stresses may be developed on each phase, due to restraints imposed by differing shrinkage rates. This internal stressing (which might be considered a type of prestressing) leads to an intermediate overall thermal expansion coefficient (Refs. 2, 3, 4).

The ultimate for enhancing thermal shock resistance through altering or selecting thermal expansion coefficients is, no doubt, a refractory with a coefficient of zero. Such materials have been developed (Ref. 5) but they do not appear to meet all the other needs of high-temperature systems. Generally, the desired characteristics of a refractory material include, besides shock resistance, a combination of good high-temperature properties and resistance to abrasion, corrosion, and frequently oxidation. The properties of BeO and SiC are such that they appear as desirable candidates for a very practical composite refractory. Both have similar moderate densities; both have similar thermal conductivities at high temperature (1500°C); BeO offers high chemical (corrosion) resistance and a high specific heat; SiC has high abrasion and corrosion resistance with a high emissivity. (The high emissivity values for SiC may be due in part to the impurity content of the common black colored material.)

The difference in thermal expansion of BeO and SiC provide an opporturity to test the calculations of Turner (Ref. 3) which indicate that these materials have favorable bulk modulus for reducing the composite coefficient of thermal expansion below that expected from a weight or volume averaging. Work done to date indicates that the measured coefficients of thermal expansion of BeO-SiC composites are even lower than those predicted by Turner's equation.

II. FABRICATION

A. EQUIPMENT

A laboratory hot press, shown in Fig. 1, was used to fabricate composite bodies from which test specimens were machined (Ref. 6). The press equipment consisted of a graphite susceptor tube placed within a transite shell. The annulus between the tube and the shell was filled with lampblack and capped with water-cooled brass plates. A water-cooled copper induction coil located external to the transite shell was powered by a 3 kilocycle motor generator (Tocco Model 64-4570-01). The furnace assembly was mounted in a steel frame fitted at the ends with hydraulic rams. Pressure was supplied to the rams from a hydraulic system built by Haskell Engineering Corp.

B. FORMING TECHNIQUES

The powders for compositing were weighed and hand-mixed in a high-alumina mortar. The initial 100-gm batch was split, providing two 50-gm charges which were loaded individually into 2-3/16-in. dia. polished graphite dies.

Five different compositions were pressed. The hot-press parameters were varied with each composition for the purpose of achieving somewhat uniform densities. It was not expected that variations in density would affect thermal expansion, since the bulk modulus of a gage is small compared to that of a solid. However, specimens were pressed from the same compositions to several different densities as a check or the possible effects of density or of the differing hot press parameters.

The temperature of the die body during forming was measured by means of a disappearing filament optical pyrometer. Pressure was measured with a precision laboratory pressure gage placed in the oil line to the hydraulic rams.

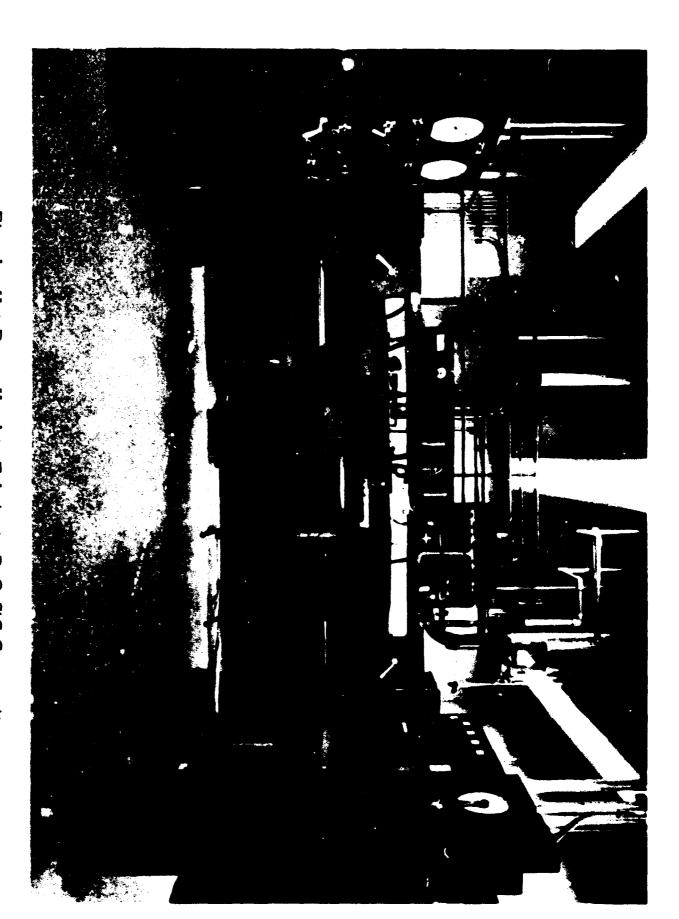


Fig. 1. Hot Press Used to Fabricate BeO-SiC Composites

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C. MATERIALS

An initial test specimen was fabricated with 25 wt % SiC polishing grit (approx. 25 micron diameter) and 75 wt % BeO (Berylco grade 1)

To more closely match the BeO particle sizes as well as the resultant grain sizes, 7-micron high-purity SiC was obtained from the Carborundum Corp. This and 7-micron "black" SiC was used for subsequent fabrications.

D. COMPOSITION AND STRUCTURE

There is no apparent reaction or solution formed between the BeO and SiC as formed at temperatures up to 1800°C, pressures to 5000 psi, and times to 4 hours. Observations were made both optically and with x-ray diffraction.

III. TEST DATA

A. THERMAL EXPANSION

The specimen wafers, as pressed, were about 1/4-in. thick by 2-3/8-in. diameter. These were cut to provide 1/8-in. x 1/8-in. x 1.575-in. specimens for thermal expansion measurement. A Leitz quartz tube dilatometer was used to obtain the linear coefficient of thermal expansion from room temperature to 1000° C. The test data is summarized in Table I.

B. DENSITY

Density values were obtained at room temperature from machined blocks by making precision measurements of volumes and weights. The results of these calculations are given in Table I.

Table I. Test Data

Specimen Number	BeO-SiC Wt. Ratio	SiC Type*	Time hrs	Hot Press Forming Temp. °C	Pressure psi	Apparent Density, gm/cm	T.D.,	Expansion Coefficient at 1000°C, cm/cm/°C × 10 ⁻⁶
144	. 75 25	В	2	1600	5000	2.52	82.4	ó. 84
145	. 50 50	В	2	1600	5000	2.42	77.7	5. 80
146	. 25 75	В	2	1800	5000	2.64	83.3	5. 19
112	. 75 25	A	4	1600	6000	3. 005av	98. 2	6. 84
120	. 50 51	A	4	1800	6000	3.015av	96.8	5. 91
125	SiC	A	4	1900	5000	2.40	74.5	4.79
126-B	.2575	A	3	1800	5000	2.96	93.1	5. 21
128	SiC	A	2	1900	5000	2.41	74.8	
132	. 50 50	A	4	1700	5000	2.89	92.7	
149	.7575	С	2	1600	5000	3.02	98.7	7.69
150	.5050	С	2	1600	5000	3.06	98.2	6.81
367-1A	BeO	-	1	1400	4000	2.29	76.1	8. 70
367-2B	BeO	-	2	1500	4000	2.75	91.4	8.60
367-2	BeO	-	3	1600	4000	2.94	97.7	8.70
367-4	. 50 50	В	4	1600	5000	2.41	77.4	6. 10
367-5	. 50 50	В	1	1500	4000	2.18	70.7	6.17
367-6	.5050	В	2	1800	5000	2.92	93.7	6.34
367-7	. 50 50	В	2	1800	5000	2.87	92. 1	o. 34
367-8	. 25 75	В	2	1800	5000	2.88	90.9	5.21
367-11	. 75 25	В	1	1500	4000	2.27	74.0	6.85
367-14	SiC	В	1.5	2000	6000	3, 12	97.0	4.55

 $^{^*}A = 25\mu$ black polishing powder

 $B = 7\mu$ light, high-purity

 $C = 7\mu \ black$

IV. DISCUSSION

The middle curve of Fig. 2 shows the linear coefficients of thermal expansion for various compositions as calculated from Eq. (1):

$$a_{r} = \frac{1^{K_{1}F_{1}/\rho_{1}} + 2^{K_{2}F_{2}/\rho_{2}}}{K_{1}F_{1}\rho_{1} + K_{2}F_{2}/\rho_{2}}$$
(1)

Where

a = the linear expansion coefficient of the aggregate

K = bulk modulus (K = $\frac{-P}{\Delta V/V} = \frac{E}{3(1-2\mu)}$, in which P is the isotropic pressure, V the volume, E the elastic modulus, and μ Poisson's ratio)

F = the weight fraction

 ρ = crystallite density

The values used for BeO and SiC are as follows:

$$\frac{\text{BeO}}{a = 8.65 \times 10^{-6} \text{ cm/cm/}^{\circ}\text{C}} \qquad \frac{\text{SiC}}{4.79 \times 10^{-6} \text{ cm/cm/}^{\circ}\text{C}}$$

$$\rho = 3.01 \text{ gm/cm}^{3} \qquad 3.25 \text{ gm/cm}^{3}$$

$$\mu = 0.204 \qquad 0.18$$

$$E = 2.6 \times \text{Kg/cm}^{2} \qquad 3.5 \times 10^{6} \text{ Kg/cm}^{2}$$

It is observed in Fig. 2 that the values calculated using Eq. (1) are displaced in a negative sense from the weight average curve. The values observed experimentally are also displaced in the same direction, but the displacement is greater. The apparent thermal expansion coefficient of BeO is significantly reduced by the addition of a SiC dispersed phase. Twenty-five weight-percent addition of SiC results in a reduction of about 10 percent in the measured expansion coefficient at 1000°C. However,

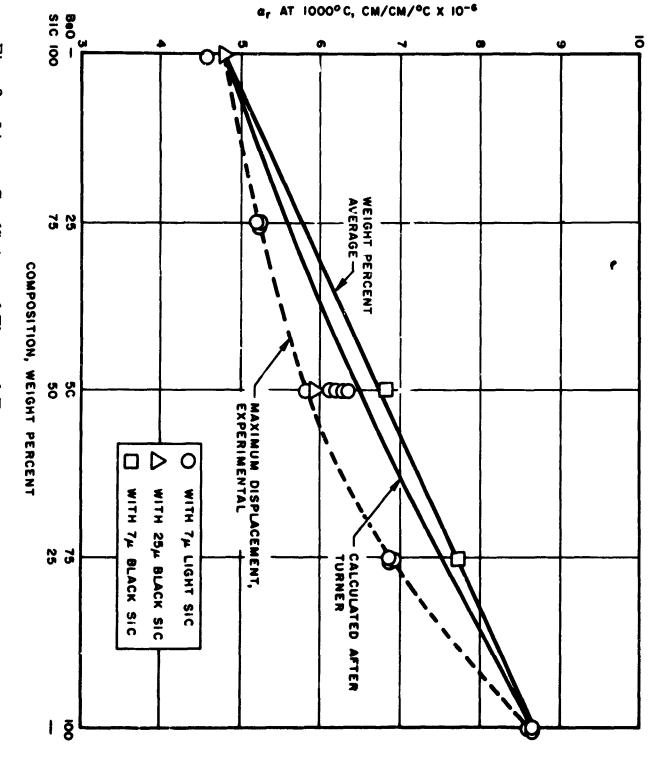


Fig. 2. Linear Coefficient of Thermal Expansion for Composite Specimens

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before this concept is directly applicable to thermal shock property enhancement, it would seem desirable to measure and optimize both strength and emissivity characteristics of the BeO-SiC composities. Continuing studies in both areas is recommended; also, strength data should be correlated with analyses of the microstructures.

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